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ELECTRON DENSITY IN AN ELECTRON BEAM
STABILIZED HIGH PRESSURE DISCHARGE

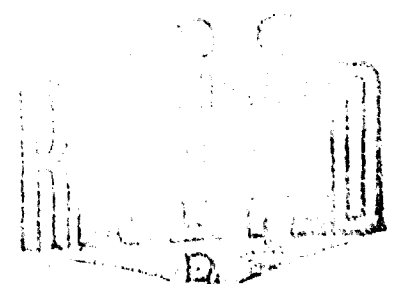
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B. B. O'Brien, Jr.

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NORTHROP CORPORATION

LASER SYSTEMS DEPARTMENT
3401 West Broadway
Hawthorne, California 90250

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PLASMA CHARACTERISTICS OF AN ELECTRON
BEAM EXCITED HIGH PRESSURE DISCHARGE

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ABSTRACT

The basic differential equations for the electron density in an electron beam stabilized discharge are formulated and a discussion of the relevant physical processes is presented. Rapid charge transfer among the positive ions is shown to result in a considerable simplification of the basic equations. An analytic solution is given which, in an approximate manner, includes the effect of oxygen attachment. A comparison between calculated electron densities and preliminary measurements is shown to be in reasonable agreement.

1.0 INTRODUCTION

Molecular gas lasers, in which the primary pumping mechanism is electron impact on the gas molecules, have shown very high efficiencies; for example up to 47% for CO¹ and 33% for CO₂.² Because of the high efficiency and relative simplicity of these electrically pumped lasers, considerable effort is being directed to the problem of scaling up the outputs of electric discharge lasers by new methods of maintaining uniform discharges in high pressure gas mixtures. One such new method is the electron beam stabilized discharge.^{3, 4, 5, 6} The calculation of the electron density of the plasma in such a discharge is the subject of this report.

An electron beam stabilized plasma differs considerably from a self-sustaining discharge in that the energy for the ionization of the gas molecules comes from a high voltage electron beam directed into the laser chamber through a thin metallic foil from a high vacuum region (see Figure 1). In the more common self-sustaining discharge, e.g., glow or radio frequency discharges, the ionization is produced by electrons generated in the plasma which possess sufficient energy. At high pressure, for example at 1 atmosphere, and at discharge times longer than about 1 μ s, such plasmas with metallic electrodes often produce low resistance local arcs which in turn cause very inhomogeneous laser pumping and unwanted heating of the active molecules. Since the ionization in an electron beam plasma is externally controlled by the energetic electron stream, one need not "heat" the electrons in the plasma to the point of causing significant ionization and thus the problem of arcing in the laser chamber is greatly reduced. Note that we distinguish between the tenuous stream of high energy electrons of the E-beam and the low energy (plasma) electrons

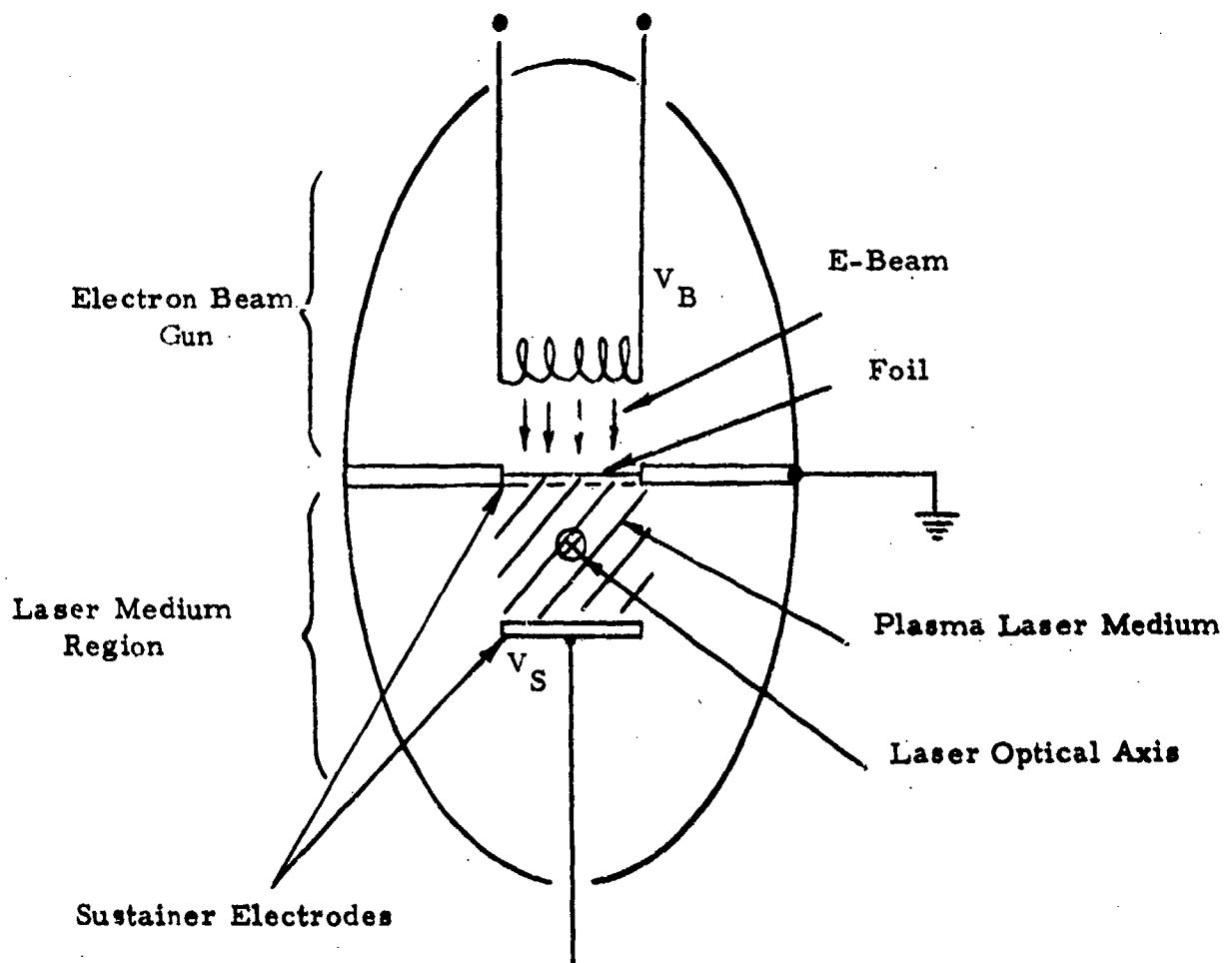


Figure 1. Cross section through the discharge region.

produced in the laser chamber by ionizing collisions between molecules and the fast beam particles. (The electron density of the beam may be about $10^6/\text{cm}^3$ and of the low energy electrons about $10^{12}/\text{cm}^3$).

Fast laser pumping rates are obtained if the average energy of the plasma electron is about 1 eV (see References 7 and 8 for CO and CO₂). This average energy is supplied by passing a current between two sustainer electrodes located in the plasma region. One electrode, which does not block the electron beam, is located near the foil and is generally at ground potential and the other electrode is maintained at the sustainer potential V_s .

The laser pumping rate is proportional to the number density of the plasma electrons, n_e , and it is the purpose of this report to discuss the physical processes which control the electron density in a multigas E-beam plasma, and to calculate n_e for some typical operating conditions. In Section 2.0 of this report the basic equations for the electron density are formulated and n_e is calculated for some exemplary cases. Also a comparison between experiment and calculation is made. A simple discussion of the basic physical processes of electron generation and loss is presented in Section 3.0. The rates for these processes are also calculated.

2.0 ELECTRON DENSITY CALCULATIONS

Molecular gas laser plasmas generally contain a mixture of gases, their positive ions (and possibly negative ions) and electrons. In electron beam stabilized plasmas the total pressure may be about one atmosphere ($\sim 10^{19}$ particles/cm³) or higher. The number densities of the ions or electrons is usually less than 10^{14} /cm³ for a total pressure of one atmosphere and thus is small compared to the neutral density. Since electron-neutral collisions are responsible for the laser pumping, with ion-neutral processes relatively unimportant, the electron density is of prime importance. Although the concentration of the various ion types is, per se, not of interest, we shall nonetheless have to consider the various ion processes since the rate of electron loss depends upon the particular ion species present in the discharge.

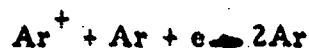
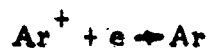
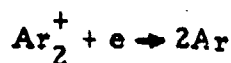
In Section 2.1 the basic set of equations which should be solved to calculate the electron density is given. For each constituent of the gas mixture, there is one first order nonlinear differential equation, and in general, the resulting set of coupled differential equations must be solved numerically. When a chemically pure gas is considered, one can write an analytic solution for n_e as a function of time (Section 2.2). One can also solve analytically an approximate equation for a single constituent gas with a small amount of oxygen (Section 2.2). Solutions for a multicomponent gas, on the other hand, are more complex. A discussion of the dominant mechanisms in typical mixtures is given in Section 2.3. Computer solutions for various gas mixtures and operating conditions are presented in Section 2.4, and in Section 2.5 some experimental measurements are compared with calculated values.

The rate constants used in the calculations of electron density are identified and reviewed in Section 3.0. Since we will use these constants in the next section, it is useful to briefly describe them here.

Electron Generation. The only electron generation process of importance is that of electron neutral collisions. We choose to divide the electrons into two classes. Class one includes the electrons of the high voltage beam and the fast secondaries which the high voltage beam produces. Class two includes the low energy electrons produced by the ionization which reside in the plasma at an energy of approximately 1 eV.

Electron Attachment. The attachment processes are two body and three body processes. A two body process, for example, is $O_2 + e \rightarrow O^- + O$, and a three body process may be illustrated by using nitrogen: $N_2 + O_2 + e \rightarrow N_2 + O_2^-$.

Recombination. The main electron-ion recombination processes are also two body and three body events, illustrated with argon:



Ion-ion recombination is the main neutralization process for negative oxygen ions. The recombination coefficient for the latter is about $10^{-6} \text{ cm}^3/\text{s}$.

Charge Transfer: The charge transfer processes are very important in determining the lifetime of the various species of ions. Rate constants given in the works of Ferguson, Schmeltekopf, Fensseld et al (cited in Section 3.5) indicate that there is rapid charge transfer from helium and nitrogen ions to carbon monoxide and carbon dioxide ions.

2.1 Basic Electron Density Equations for a Multicomponent Gas. To formulate a basic set of equations for a multicomponent gas we shall assume quasi-neutrality and homogeneity. Quasi-neutrality implies near equality between free positive and negative charges, and homogeneity implies that the temporal rate of change of ion number density is governed only by ion loss and generation rates. Let $n(j^+)$ represent the number density of positive ions of the j th species, n_e the electron density, and $n(O_2^-)$ the number density of charged oxygen molecules as the only negative ions present. From the quasi-neutrality requirements, we can write

$$n_e = \sum_j n(j^+) - n(O_2^-) \quad (1a)$$

and from the homogeneity concept we write (for each particle species)

$$dn(j^+)/dt = G(j^+) - L(j^+) \quad (1b)$$

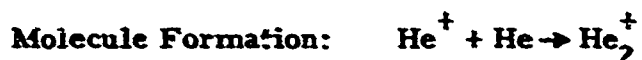
and

$$dn(O_2^-)/dt = G(O_2^-) - L(O_2^-) \quad (1c)$$

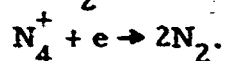
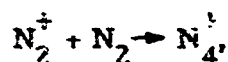
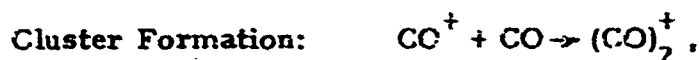
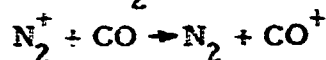
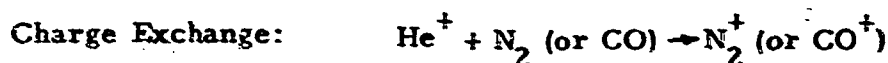
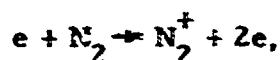
where the G and L terms refer to the generation and loss rates for each type of ion.

There are many possible reactions in a typical laser gas which must be included in the loss and generation terms of Eqs. (1b) and (1c). In order to obtain a tractable set of equations we shall confine our attention to those reactions listed below and discussed in Section 3.5.

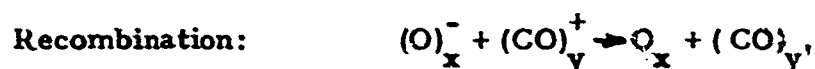
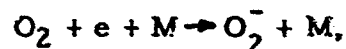
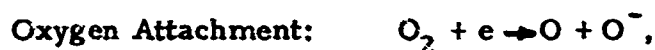
For a pure gas, for example helium, the dominant reactions for our purposes are:



For a gas mixture, for example helium, nitrogen and carbon monoxide, the dominant reactions are:



For a gas mixture containing a small amount of oxygen, in addition to the above, we have:



where at this stage of the investigation the specific ions and molecules, designated by the subscripts x and y , cannot clearly be identified and M is any atom or molecule.

The charge exchange processes, schematically indicated above, enter into the ion density rate equations (1b) as a loss for the initial ion species and a generation for the resultant species.

The basic equations (1a, b and c) can be simplified by considering the approximate rates of the various processes. In particular, the average lifetime for helium or nitrogen ions, considering charge transfer processes, is in the order of 10^{-10} sec, and the lifetime, determined by recombination processes, is in the order of 10^{-5} to 10^{-7} sec. Since we are not interested in the ion density but rather electron density, and since the free electrons are not involved in the charge transfer process, we can simplify the basic equations. We sum up the effect on the basic equations of the dominant reactions given above as follows (neglecting for the moment, any attachment):

- (a) Helium, nitrogen and carbon monoxide ions (and electrons) are generated as indicated above.
- (b) Helium and nitrogen ions charge transfer to carbon monoxide ions infinitely fast.
- (c) Because of (b), the effective generation rate for the carbon monoxide ions is the sum of all the generation rates.
- (d) The time to form $(\text{CO})_2^+$ or $(\text{CO})_x^+$ is estimated to be about 10^{-9} sec. Since this time is short compared to the recombination time, we may assume that this process proceeds arbitrarily fast.
- (e) The recombination of $(\text{CO})_2^+$ or $(\text{CO})_x^+$ indicated above is then the only important loss process for electrons.

Although the electron-ion recombination process for carbon monoxide (and other ions) involves a larger molecule, i. e., $(\text{CO})_x^+$ we shall simplify our

notation, whenever possible, by writing CO^+ instead of $(\text{CO})_x^+$. We shall use this convention for other ions as well.

The recombination rate for carbon monoxide ions is $\alpha(\text{CO}^+) n(\text{CO}^+) n_e$ where $\alpha(\text{CO}^+)$ is the recombination rate constant for CO^+ . For the case of no oxygen, $n(\text{CO}^+) = n_e$, and the recombination rate reduces to $\alpha(\text{CO}^+) n_e^2$. Using this loss term, Eqs. (1a) and (1b) reduce to

$$dn_e/dt = G(\text{CO}^+) + G(\text{He}^+) + G(\text{N}_2^+) - \alpha(\text{CO}^+) n_e^2. \quad (2)$$

It is also assumed that the relative concentration of oxygen atoms, when present, is less than about 10^{-2} , and therefore the number of electrons derived from positive oxygen ionization is negligible. The effect, however, of even a small amount of oxygen on electron loss through attachment, may be severe, and the coupled set of Eqs. (1a, b and c) must then be solved.

The statements (a) and (d) are unaffected by oxygen and they still serve to simplify Eq. (1b). In Eq. (1c), $G(\text{O}_2^-)$ represents the attachment process and $L(\text{O}_2^-)$ the ion-ion recombination process. We shall denote the oxygen recombination coefficient with $\alpha(\text{O}_2^-)$. The simplified set of equations for a helium, nitrogen, carbon monoxide and oxygen mix is:

$$n_e = n(\text{CO}^+) - n(\text{O}_2^-) \quad (3a)$$

$$\begin{aligned} \frac{dn(\text{CO}^+)}{dt} = & G(\text{CO}^+) + G(\text{He}^+) + G(\text{N}_2^+) \\ & - \alpha(\text{CO}^+) n_e n(\text{CO}^+) - \alpha(\text{O}_2^-) n(\text{O}_2^-) n(\text{CO}^+) \end{aligned} \quad (3b)$$

$$\frac{dn(\text{O}_2^-)}{dt} = n_e \beta' - \alpha(\text{O}_2^-) n(\text{O}_2^-) n(\text{CO}^+) \quad (3c)$$

where β' (discussed in Section 3.4) is the attachment rate constant. We have ceased distinguishing between O^- and O_2^- (or other negative oxygen ions).

Because of the very anisotropic electron velocity distribution, it is convenient to divide the electrons into two classes, as indicated earlier, and to divide the ionization function $G(j^+)$ into two parts. Thus, $G(j^+)$ is written as

$$G(j^+) = i n(j) g(j) + n_e n(j) F(T_e, j) \quad (4)$$

where the first term on the right hand side corresponds to the ionization caused by the electron beam and the second term the ionization caused by the electrons having random energies of the order of 1 eV. $n(j)$ is the molecular or atomic number density of species j , i is the electron beam current, and $g(j)$ is a normalized ionization rate for species j . $F(T_e, j)$ is an ionization rate per molecule or atom, which depends on the electron energy. It shall be assumed that the random motion of the electrons can be approximated by a Boltzmann-Maxwell distribution* at temperature T_e . The functions $g(j)$ and $F(T_e, j)$ are discussed in Sections 3.1 and 3.2 respectively.

* Although Nighan^{8,9} has calculated that the electron velocity distribution in CO_2 and CO gas mixtures for the usual discharge conditions will depart from a Maxwellian, we will often treat this approximately isotropic random motion as thermal.

2.2 The Analytic Solution of Electron Density for a Single Component

Gas. In this section the basic equation for the electron density (Eq. 1) will be simplified by considering only one gas, for which an analytic solution can be obtained. An approximate equation that includes the effects of oxygen will also be derived and solved.

For this example we shall assume that the fill gas is argon with a small amount of oxygen. Since the percentage of oxygen is assumed small, we neglect the positive ions of oxygen. Eqs. (1a, b and c) now reduce to:

$$n_e = n(\text{Ar}^+) - n(\text{O}_2^-) \quad (5a)$$

$$\begin{aligned} \frac{dn(\text{Ar}^+)}{dt} = & n(\text{Ar})ig(\text{Ar}) + n_e n(\text{Ar}) F(T_e, \text{Ar}) \\ & - \alpha(\text{Ar}^+)n(\text{Ar}^+)n_e - \alpha(\text{O}_2^-)n(\text{O}_2^-)n(\text{Ar}^+) \end{aligned} \quad (5b)$$

$$\frac{dn(\text{O}_2^-)}{dt} = \beta' n_e - \alpha(\text{O}_2^-)n(\text{O}_2^-)n(\text{Ar}^+) \quad (5c)$$

This set of equations cannot be solved simply without numerical methods.

We shall obtain analytical solutions for three cases: (a) no thermal generation ($F(T) = 0$) and no oxygen attachment ($\beta' = 0$), (b) thermal generation but no attachment, and (c) no thermal generation but attachment. For cases (a) and (b), we obtain simple solutions to the exact equations. For (c) we must approximate Eq. (5) in order to solve for n_e . We shall assume that the electron beam is on for a length of time p . Figure 2 shows the value of the electron density as a function of time for the cases (a), (b) and (c).

Case (a): For $F(T_e) = 0$ and $\beta' = 0$ we have

$$\frac{dn_e}{dt} = -an_e^2 + c \quad (6a)$$

where $a = \alpha(\text{Ar}^+)$ and $c = n(\text{Ar})ig(\text{Ar})$

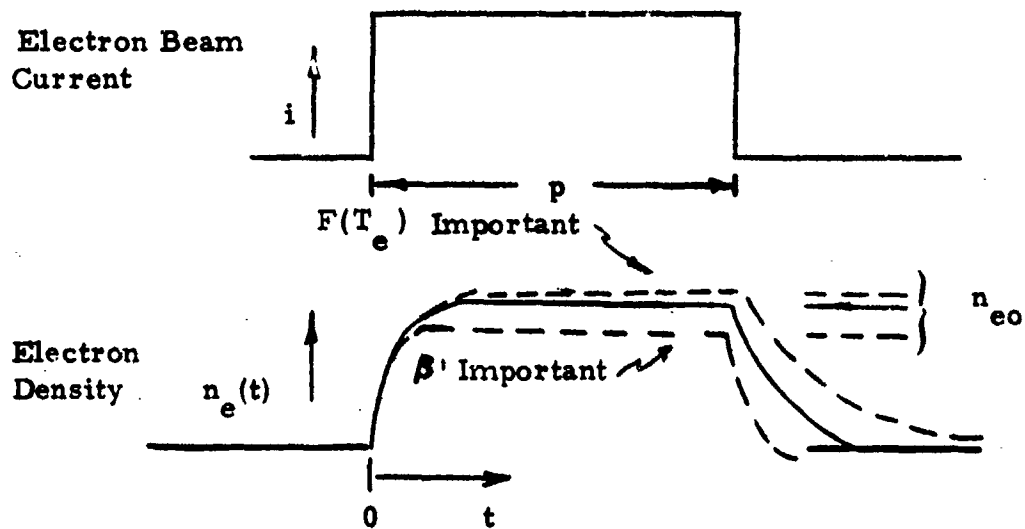


Figure 2. Electron beam current and electron density vs time.

The solution is:

$$\text{For the buildup of } n_e: n_e = (c/a)^{1/2} \tanh \sqrt{ac} t \quad 0 \leq t \leq p \quad (6b)$$

$$\text{For the decay of } n_e: n_e = (a(t - p) + 1/n_{eo})^{-1} \quad p < t \quad (6c)$$

where $n_{eo} = n_e(t = p)$.

Eq. (6b) and (6c) are plotted in Figure 2.

If we define the characteristic rise and decay times to be $\tau_r = (ac)^{-1/2}$ and $\tau_d = (n_{eo} a)^{-1}$ respectively, then we note that $\tau_r = \tau_d$ if the build up transient is complete (i. e., if $n_{eo} \cong (c/a)^{1/2}$). A sample calculation for 760 torr argon with 1 mA/cm² electron beam current yields a steady state equilibrium electron density, n_{eq} , of 4.3×10^{12} /cm³ and a rise time of 1 μ s. This was calculated using $a = 8.8 \times 10^{-7}$ cm³/s and $c = 1.2 \times 10^{18}$ (cm³ s)⁻¹.

Case (b): For $F(T_e) \neq 0$ and $\beta' = 0$ we have

$$dn_e/dt = -a n_e^2 - b n_e + c \quad (7a)$$

where we define $b = \beta' - n(Ar)F(T_e)$. In this case b simplifies to $-n(Ar)F(T_e)$. The solution for this case is:

$$\text{For the buildup of } n_e: n_e = \frac{2c(1 - e^{-tR})}{(-b + R) e^{-tR} + b + R}, \quad 0 \leq t \leq p \quad (7b)$$

$$\text{For the decay of } n_e: n_e = \frac{n_{eo}}{\left(\frac{an_{eo}}{b} - 1\right) e^{b(t-p)} - \frac{an_{eo}}{b}}, \quad p < t \quad (7c)$$

where $R = (b^2 + 4ac)^{1/2}$.

Case (c): For this case Eqs. (2a, b, and c) can be combined to yield

$$\frac{dn_e}{dt} = -an_e^2 (1 + n(O_2^-)/n_e) - bn_e + c. \quad (8)$$

Considering attachment and $n(O_2^-)/n_e \ll 1$, Eq. (8) is of the form of (7a) and its solution is again given by (7b) and (7c), but the coefficient $b = \beta'$ for this case.

The value of $n(O_2^-)/n_e$ can be easily estimated for steady state conditions. From Eq. (5c) one calculates

$$n(O_2^-) = [\beta' / \alpha(O_2^-)] (1 + n(O_2^-)/n_e)^{-1} \quad (9)$$

which, for $n(O_2^-)/n_e$ small compared to one, gives $n(O_2^-) \approx \beta' / \alpha(O_2^-)$. For typical values of $\beta' = 5 \times 10^3 s^{-1}$ and $\alpha(O_2^-) = 10^{-6} cm^3 s^{-1}$, one calculates $n(O_2^-) = 5 \times 10^9 cm^{-3}$. For $n_e = 10^{12} cm^{-3}$ the above approximation to Eq. (8) for steady state conditions is well justified. During the initial build up of n_e , the ionization term c is more important than the electron recombination term and therefore the approximation to Eq. (8) should also be good for this period. Late in the after-glow, the recombination term is less important than the attachment term and again the approximation to Eq. (8) should cause no significant error in calculating n_e for this period.

If the turn on transient is complete and steady state is achieved, the equilibrium electron density, n_{eq} for cases (b) and (c) is

$$n_{eq} = [-b + (b^2 + 4ac)^{1/2}] (2a)^{-1}. \quad (10)$$

The solution for n_e for cases (a), (b), and (c) is shown in Figure 2. The initial rise in electron density is governed solely by the electron beam ionization rate. The final phase of the electron density build-up is only slightly influenced by attachment and thermal ionization; it proceeds faster for the case of attachment and slower for the case of thermal generation. The rate of decay of the electron density is strongly influenced by attachment and thermal generation as shown in Figure 2.

2.3 The Electron Density Function for a Multicomponent Gas. The simplified basic equations (3a, b and c) for a gas mixture are of the same form as those for a single component gas (5a, b and c). Therefore, the solutions for n_e in equations (7b, c) and (9), also represent analytic solutions for the multicomponent gas case, provided that the definition of the constants a , b , and c is generalized. It should be noted that the differential equation (7a) is approximate when electron attachment is significant, since $n(j^+)$ is not equal to n_e .

For the multicomponent gas, the coefficients a , b , and c to be used in the approximate solution (7b, c) are:

$$a = \alpha(j^+) \quad (11a)$$

where (j^+) is the final ion type in the rapid charge transfer process, e.g., CO^+ or CO_2^+ ,

$$b = \beta' - \sum_j n(j)F(T, j) \quad (11b)$$

where β' , derived in Section 3.4, includes the 2 body and 3 body attachment process and $n(j)F(T_e, j)$ is the thermal ionization rate of atom species j , and

$$c = i \sum_j n(j)g(j) \quad (11c)$$

where $i n(j)g(j)$ is the rate of ionization produced by the electron beam in species j .

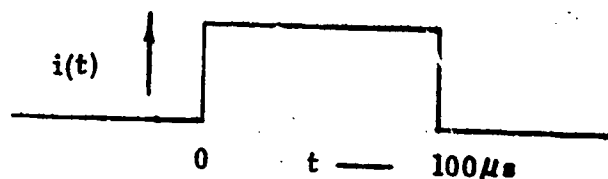
The qualitative behaviour of n_e is depicted in Figure 3 for various cases. An electron beam pulse of 100 μs duration is assumed and results for pure and multicomponent gases with and without attachment and thermal generation are shown. In the next section, numerical solutions of Eqs. (3a, b, c) are presented and discussed. In the section following, comparisons are made between the calculated and measured results.

2.4 Solutions for Various Operating Conditions. Computer solutions of n_e for various operating conditions are presented in this section. A 100 keV electron beam pulse of 100 μs duration and a total gas pressure of 760 torr at 273°K was assumed for each case. Table I lists the cases considered with parameters chosen to illustrate the effect on the electron density caused by:

- Changing the beam current; Figure 4c
- Including attachment; Figures 4d, 6b and 7b,
- Including thermal generation; Figures 4e, 6c and 7c,
- Changing gas; Figures 5a, b, c, and d, and
- Using different laser gas mixtures; Figures 6 and 7.

With no attachment or thermal generation the effect of changing the beam current is as anticipated: the maximum electron density varies as the square root of the current, and the rise and fall times decrease at higher currents. From Eq. (4) one notes that a change in the gas density is equivalent to a change in the beam current.

The electron beam



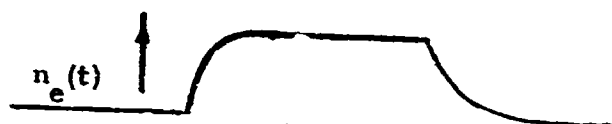
For pure CO with no thermal generation. (High generation and recombination rates)



For pure He with no thermal generation. (Low generation and very low recombination rates.)



For CO, N₂, and He with no thermal generation and attachment.



For CO, N₂ and He with thermal generation but no attachment.



For CO, N₂, and He with attachment but no thermal generation.



Figure 3. Effects of various basic processes on electron density.

TABLE I - Table of Rates used in Figures 4 - 7

| Figure | Gas Pressure (Torr at 273°K) | α (cm ³ /s) | E-Beam mA/cm ³ | $i \Sigma n(i)g(i)$ (cm ³ /s)-1 | $\Sigma n(j)E(T_e, j)$ (cm/s) ⁻¹ e.j) | β (s ⁻¹) |
|--------|--|----------------------------------|------------------------------|---|---|----------------------------|
| 4a | N ₂ 760 | 8 x 10 ⁻⁸ | 0.01 | 0.076 x 10 ¹⁷ | 0 | 0 |
| 4b | N ₂ 760 | 8 x 10 ⁻⁸ | 1.0 | 7.6 x 10 ¹⁷ | 0 | 0 |
| 4c | N ₂ 760 | 8 x 10 ⁻⁸ | 100 | 760 x 10 ¹⁷ | 0 | 0 |
| 4d | Same as 4b but with 4.0 torr of oxygen | | | | | 9 x 10 ⁴ |
| 4e | Same as 4b but with thermal generation T _e = 1.1 eV | | | | 2.7 x 10 ⁴ | |
| 5a | He 760 | 4 x 10 ⁻⁹ | 1.0 | 1.9 x 10 ¹⁷ | 0 | 0 |
| 5b | N ₂ 760 | 8 x 10 ⁻⁸ | 1.0 | 7.6 x 10 ¹⁷ | 0 | 0 |
| 5c | CO 760 | 1 x 10 ⁻⁷ | 1.0 | 7.6 x 10 ¹⁷ | 0 | 0 |
| 5d | CO ₂ 760 | 1 x 10 ⁻⁷ | 1.0 | 14 x 10 ¹⁷ | 0 | 0 |
| 6a | { He 380 N ₂ 253 CO 127 } | 1 x 10 ⁻⁷ | 1.0 | 4.75 x 10 ¹⁷ | 0 | 0 |
| 6b | Same as 6a but with 2.0 torr of oxygen | | | | 0 | 2.7 x 10 ⁴ |
| 6c | Same as 6a but with thermal generation T = 1.0 eV | | | | 2.9 x 10 ⁴ | 0 |
| 6d | { N ₂ 506 CO 254 } | 1 x 10 ⁻⁷ | 1.0 | 7.6 x 10 ¹⁷ | 0 | 0 |
| 7a | { He 380 N ₂ 253 CO ₂ 127 } | 1 x 10 ⁻⁷ | 1.0 | 5.75 x 10 ¹⁷ | 0 | 0 |
| 7b | Same as 7a but with 2.0 torr of oxygen | | | | | 2.7 x 10 ⁴ |
| 7c | Same as 7a but with thermal generation T = 1.0 eV | | | | 4.2 x 10 ⁴ | 0 |

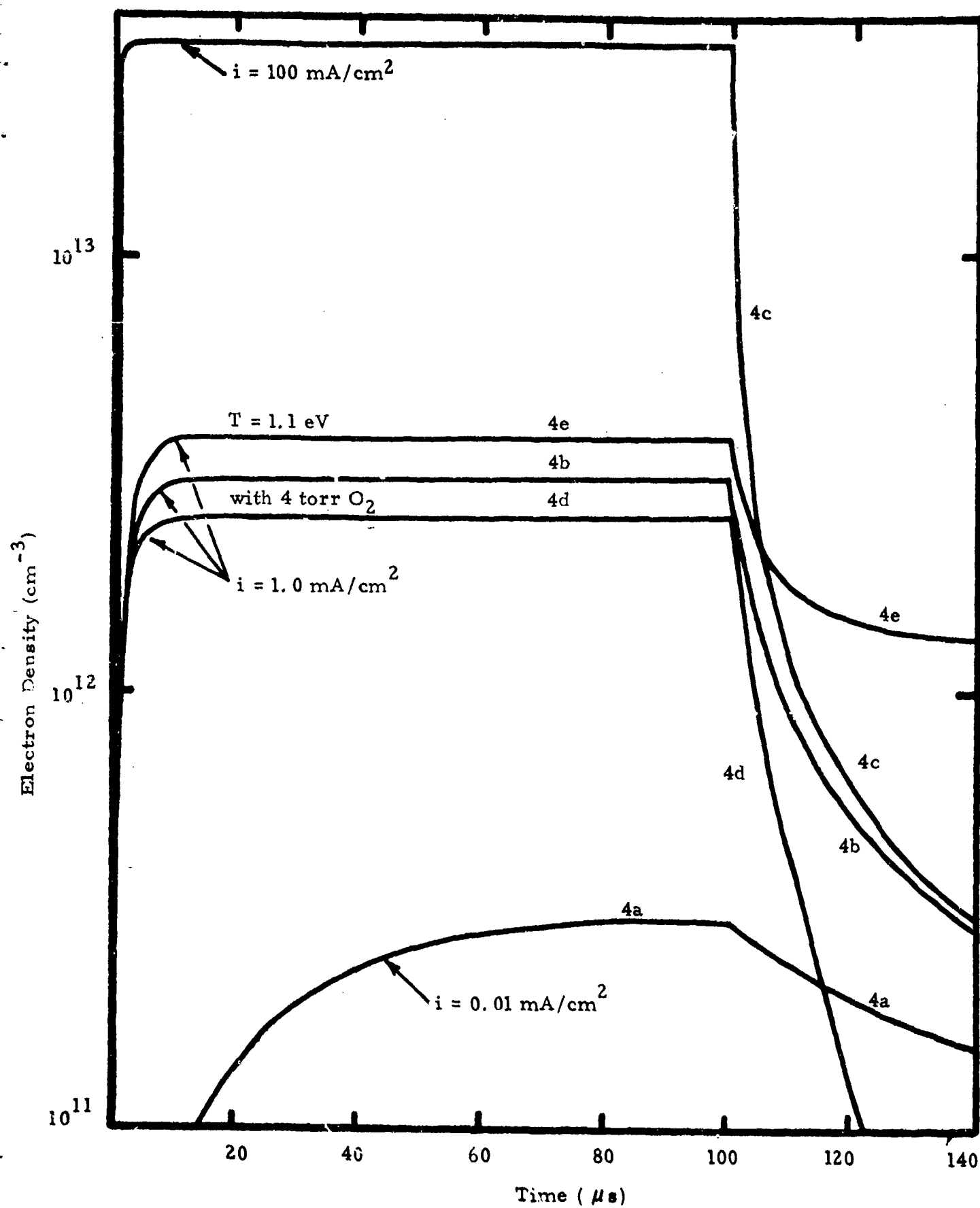


Figure 4. Electron Density for Nitrogen under Various Conditions.

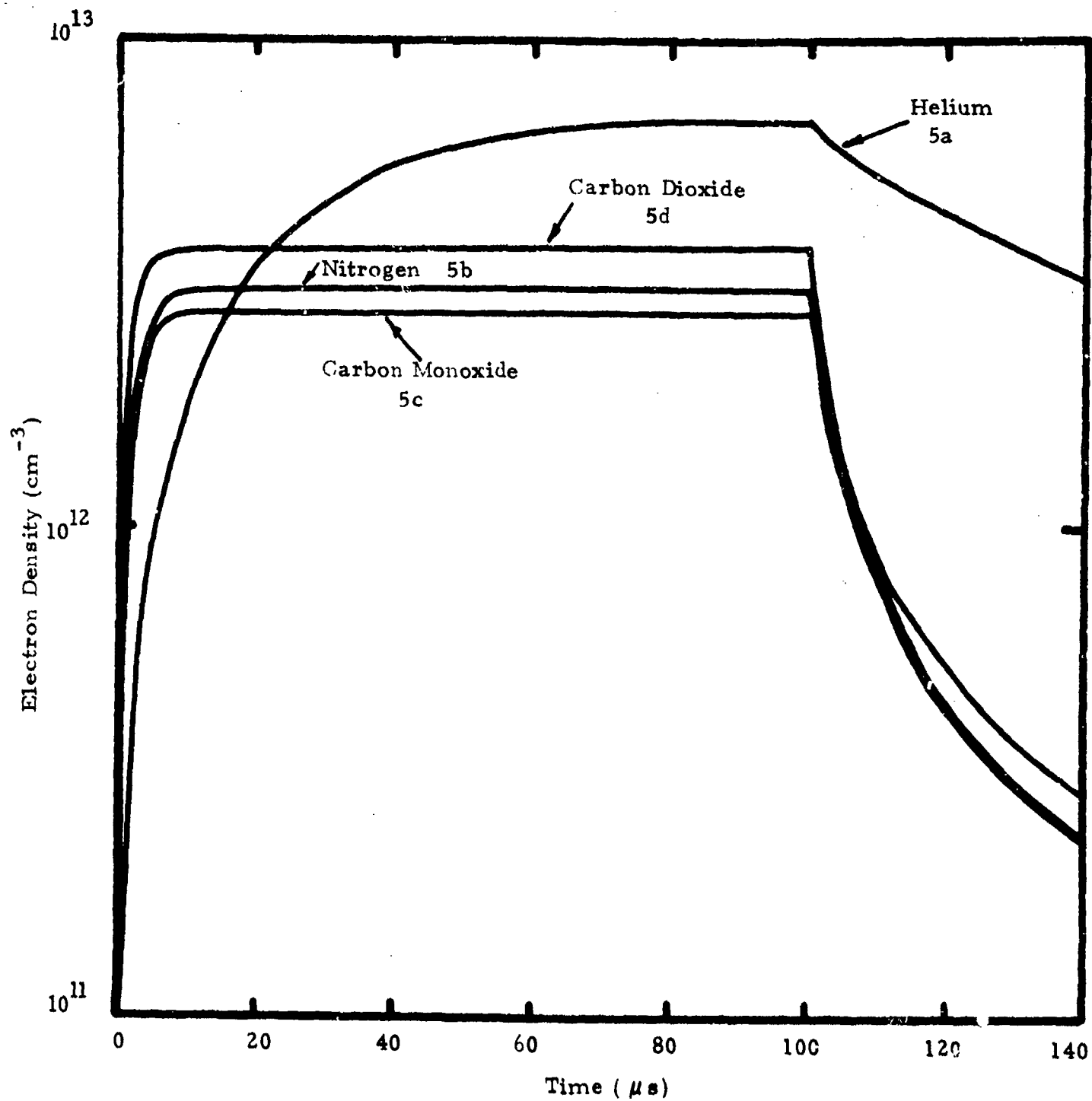


Figure 5. Electron density versus time for 760 torr of fill gas.

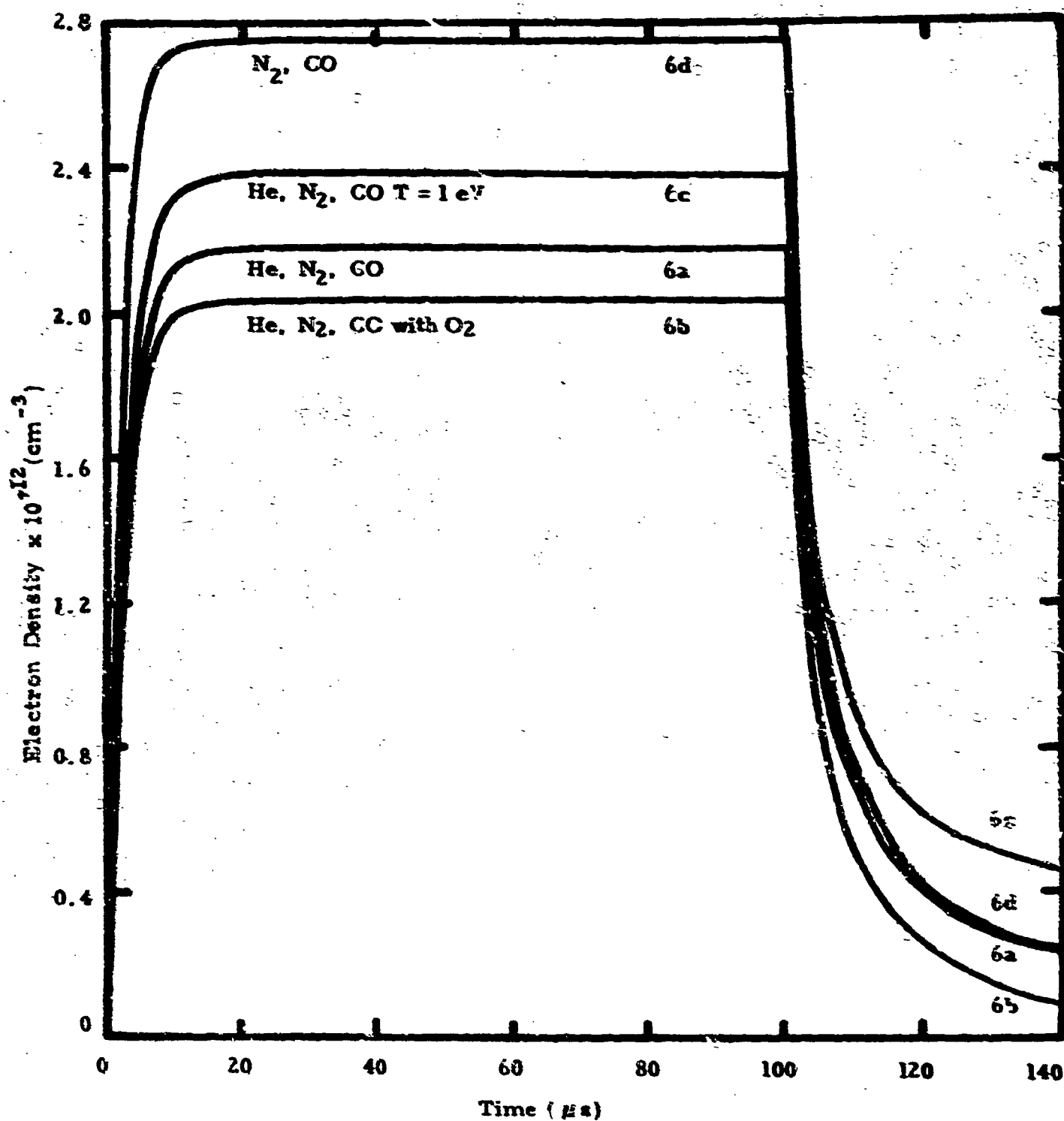


Figure 6. Electron Density for CO Mixtures.

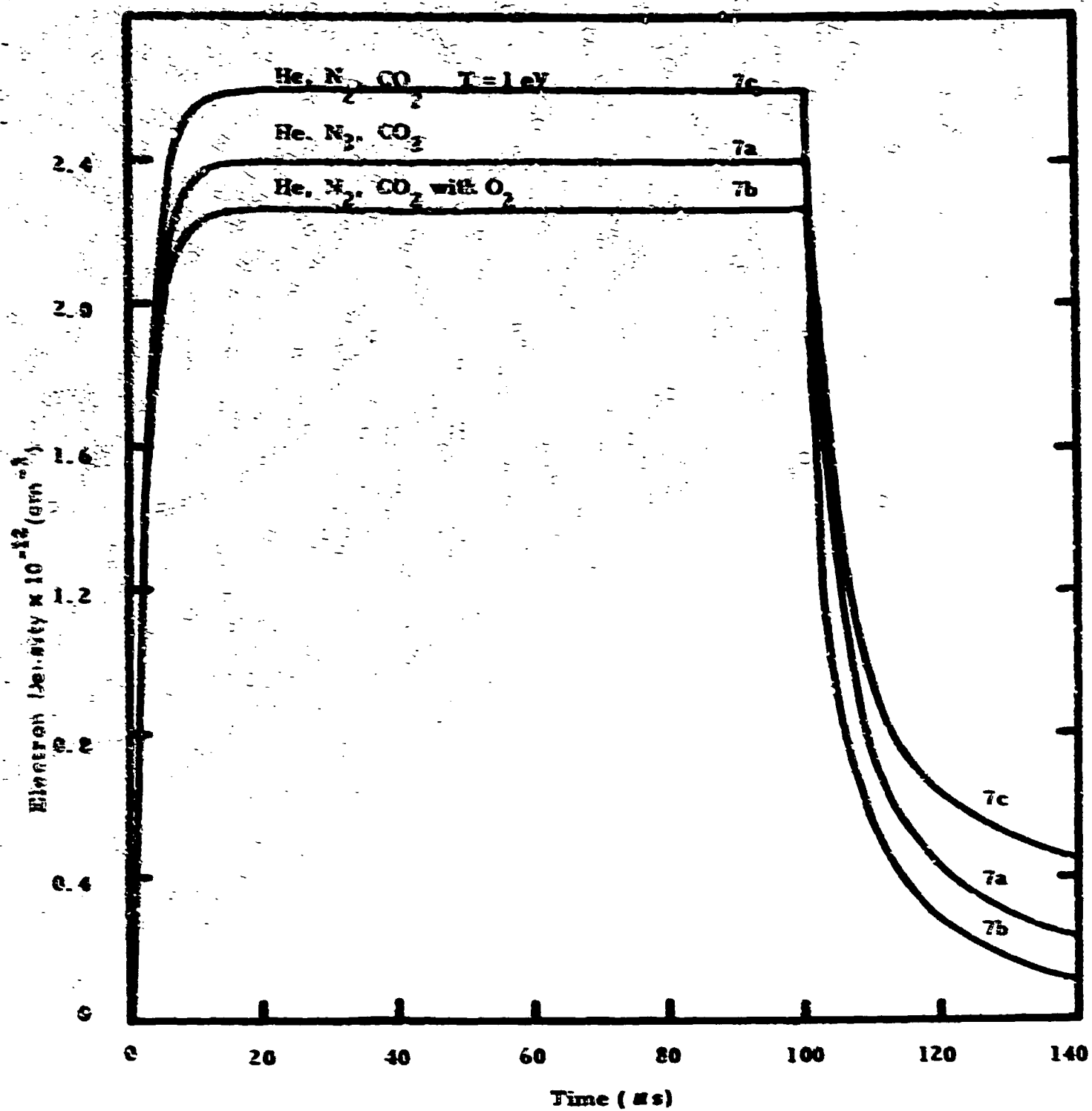


Figure 7. Electron Density for CO₂ Mixtures.

In Figure 5 the effect of the low generation rate and the very low recombination rate in helium is noted; namely long rise and fall times but a high steady state electron density (its value would be $7 \times 10^{12}/\text{cm}^3$ for a sufficiently long pulse). The formation of negative ions is to be expected for CO and CO₂ discharges, but it is not considered in Figures 5c, and d.

Figures 5 and 7 give the electron density for typical laser gas mixtures. Whenever helium is present, there is rapid charge transfer to carbon monoxide or carbon dioxide. Since the generation rate of helium is low compared to N₂, CO or CO₂, there is only a small contribution to the electron density from the helium (compare Figures 6a and 6d).

The purity of CO is often a problem because carbonyls are often present and they attach electrons rapidly.¹⁰ The inclusion of some oxygen attachment should approximate the effect of carbonyl electron attachment. Little is known about the recombination of carbon dioxide because, in most discharges, many of the free electrons attach and form negative ions which mask the recombination process. (CO₂ has a large cross section for negative ion formation at electron energies of about 4 eV).¹¹

2.5 Comparison of Measured and Calculated Currents. In electrical discharges, operating at atmospheric pressure with a low degree of ionization, it is difficult to measure the electron density directly (by probes or by microwave methods). A measure of the accuracy of the electron density calculations may be obtained by comparing the current which flows between the sustainer electrodes in the laser plasma region (see Figure 1). The expected value of current density i_s is

$$i_s = |en_e v_d| \quad (12)$$

where e is the electron charge and v_d the electron drift velocity in the electric field of the sustainer electrodes. The values for v_d were obtained from Brown,¹² and the electric field was taken to be the voltage between the electrodes divided by the separation. The total measured sustainer current divided by the area gave the average current density i_a .

The results of preliminary measurements of discharge currents in nitrogen and in carbon monoxide are shown in Figures 8 and 9. The electron beam current density was about 0.5 mA/cm^2 , and the current measurements in the laser cavity were made after the electron density build up was complete. Both β' and $F(T_e)$ were taken to be zero in the calculation. The cathode fall, estimated to be about 400 volts, was neglected.

The agreement between the measured and calculated discharge current values for nitrogen was better than expected with this crude model. The lower measured current values for carbon monoxide may be due to some attachment process.

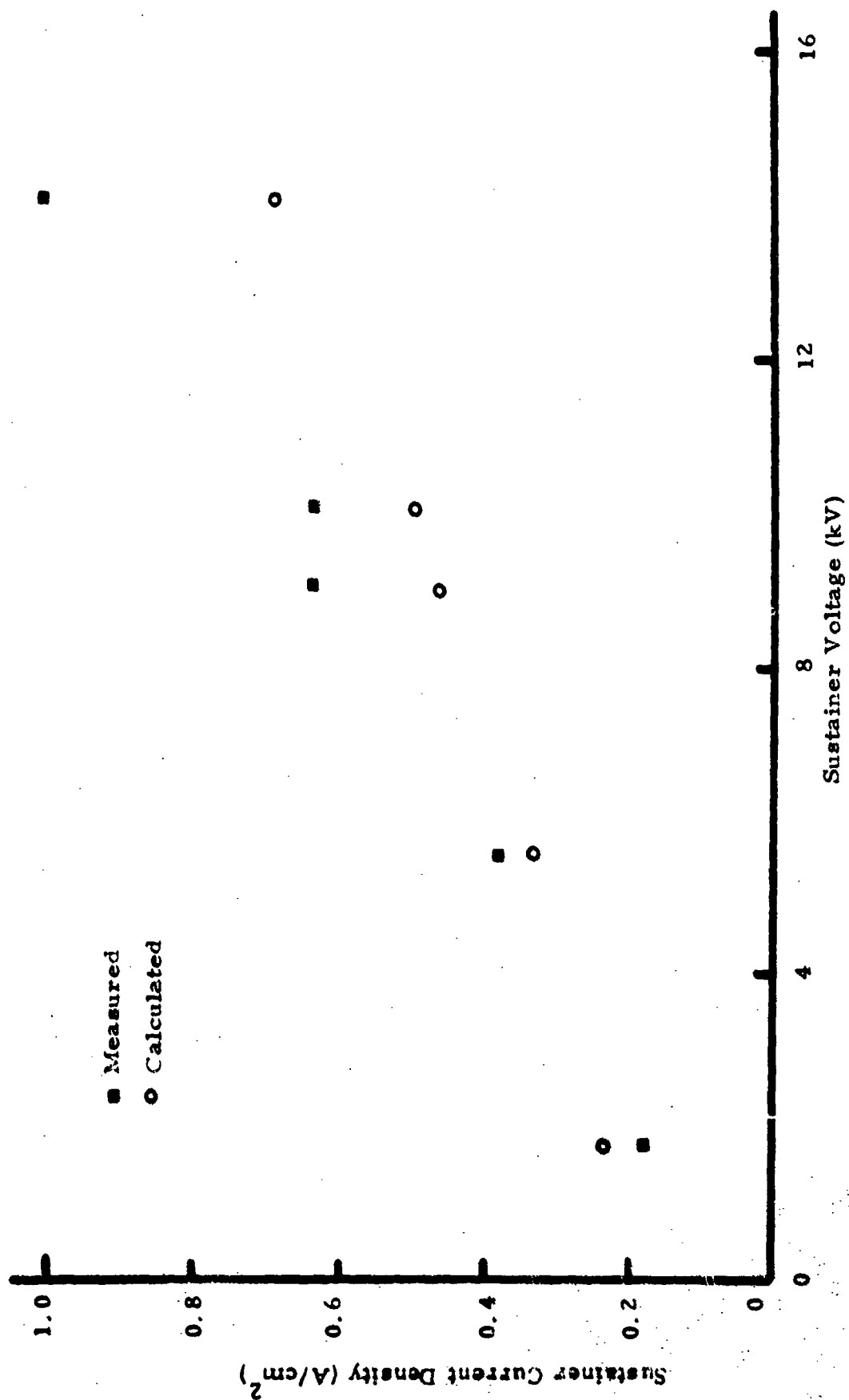


Figure 8. Sustainer Voltage versus Sustainer Current Density (760 torr N₂, i E-Beam \approx 0.55 mA/cm²)

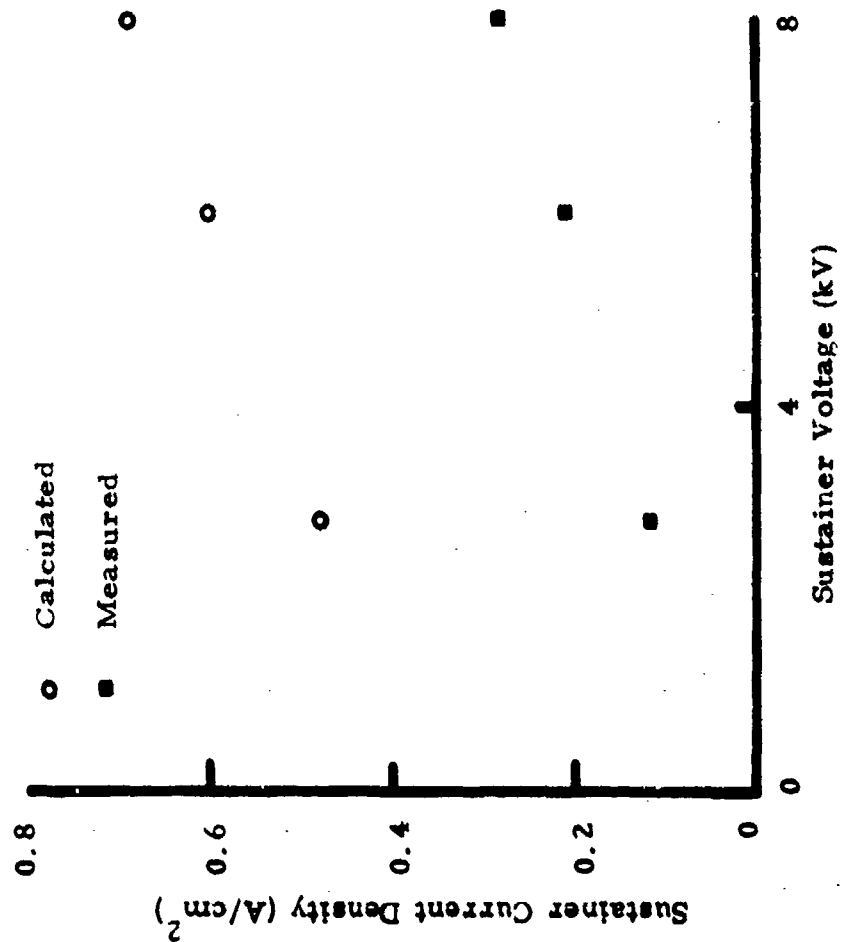


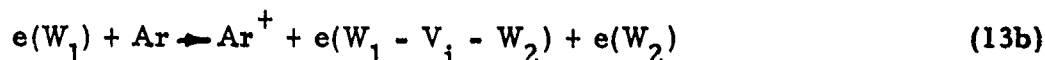
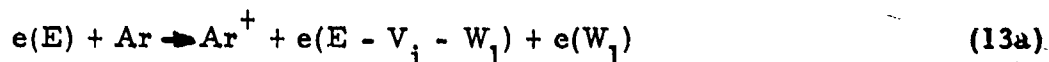
Figure 9. Sustainer Voltage versus Sustainer Current Density
(760 torr CO, i E-beam \approx 0.55 mA/cm²)

3.0 BASIC PHYSICAL PROCESSES

In this section we shall discuss the basic reactions and the reaction rates giving rise to:

- (1) Ionization of the fill gas atoms by the electrons of the E-beam,
- (2) Ionization of the atoms by electrons having about 1 eV thermal energy,
- (3) Electron-ion and ion-ion recombination,
- (4) Electron attachment to and detachment from oxygen, and
- (5) Charge transfer processes.

3.1 Ionization of Fill Gas Atoms by Electrons of the E-Beam. The reactions, by which the very energetic electrons from the electron gun ionize neutral gas particles, are cascade processes, which may be expressed as follows using Ar.



where E is the energy of the beam electron ($E \sim 100$ keV), V_i is the ionization energy and W is the kinetic energy of the ejected electron.

The energetic electron ionizes an argon atom and loses an amount of energy approximately equal to the ionization energy plus the kinetic energy, W_1 , of the secondary electron. The cross sections σ_i for this process, listed in Table II, are discussed below. The generation process, however, is not complete as the secondary electron at energy W_1 may then ionize additional atoms. The likelihood of this latter process is more difficult to calculate. The highly energetic electron may also impart some further energy to the argon atom, however, this is not included in the above equations.

TABLE II. Ionization Rates, Cross Sections, and Stopping Power

| Gas | $\frac{g}{(s \cdot mA/cm^2)}$ | Stopping Power $eV/cm \text{ at } 3.5 \times 10^{16} \text{ cm}^{-3} \text{ density}$ | σ_i Primary Ionization $(cm^2) \text{ at } 100 \text{ keV}$ |
|-----------------|-------------------------------|--|---|
| He | 0.007 | 1.1 | 0.23×10^{-18} |
| Ne | 0.025 | 4.0 | 0.74 |
| Ar | 0.045 | 6.86 | 1.90 |
| Kr | 0.087 | 12.2 | 3.1 |
| Xe | 0.13 | 17.0 | 4.95 |
| H ₂ | 0.005 | 1.0 | 0.40 |
| N ₂ | 0.028 | 6.0 | 1.7 |
| O ₂ | 0.037 | 6.8 | 2.0 |
| CO | Assume same as N ₂ | | |
| CO ₂ | 0.051 | 9.8 | |

We will first discuss the cross section calculation for process (13a) and the distribution of kinetic energy (W_1) of the ionized electron, and then compare these energy losses of the primary electron with stopping power data. We will then discuss process (13b) and denote with ϵ the number of further ionizations caused by the electron at energy W_1 . Finally, we will relate the total number of ion pairs/cm caused by the passage of one high energy electron to the results obtained by a rule of thumb calculation. The latter is based on calculating the ion pair generation rate from the stopping power by assuming that each ion pair takes about 30 - 35 eV energy of the primary electron.

The cross sections for process (13a) have been measured by many investigators¹³ at low energies (≤ 1 keV). Less work has been done at energies around 100 keV. Schram et al¹⁴ have made measurements of the first eight gases listed in Table II at energies up to 20 keV and have tabulated the constants necessary to use the Bethe formula,¹⁵ which allows one to calculate the ionization cross sections for 100 keV primary electrons (or other energies of interest).

Using the very simple collisional model presented in reference 15, (page 276), which considers a highly energetic electron encountering other electrons at rest (process 13a), one can calculate σ_i and the average kinetic energy of the ejected electron $\langle W_1 \rangle$. For $E = 100$ keV and $V_i = 15$ eV, one calculates $\sigma_i = 0.043 \times 10^{-18} Z \text{ cm}^2$ and $\langle W_1 \rangle = 105$ eV, where Z is the atomic (or molecular) number. The calculated value of σ_i is about a factor of two too small when compared with measured values in Table II. The value of $\langle W_1 \rangle$ is within about 20% of the value one obtains from stopping power data. For example, in argon at 1 atmosphere, with

$\sigma_i = 1.9 \times 10^{-18} \text{ cm}^2$, there are 51 direct ionizing processes per cm for each primary electron. The energy loss per cm for each primary electron is thus 51 ($V_i + \langle W_i \rangle$) = 6.1 keV/cm. The tabulated energy loss given by Berger and Seltzer¹⁶ is 5.1 kV/cm.

The kinetic energy of the ejected electron decays from a value of W toward a steady state value ($\sim 1 \text{ eV}$). There are many processes which compete for this excess kinetic energy and we will not attempt to discuss how this energy is split between ionizing and nonionizing processes. Fano¹⁷ proposed a theory which states that the average energy, w , which is expended in producing an ion pair by the processes of Eq. (13a) and (13b), and subsequent ionization processes, should be quite independent of the atom structure and is about 30 eV. This is born out experimentally and more precise values, taken from reference 15 (page 233), are given below. Also shown below are some measured values of ϵ , the number of further ion pairs produced by the electron ejected in the primary ionization process, Eq. (13a).

| | <u>w(ev)</u> | <u>ϵ¹⁸</u> |
|-----------------|--------------|---|
| Hydrogen | 35 | 1.3 |
| Nitrogen | 36.2 | 2.3 |
| Argon | 26 | 2.7 |
| Oxygen | 32.3 | 2.5 |
| Helium | 30 | |
| Neon | 28 | |
| Krypton | 24 | |
| Xenon | 22 | |
| CO | 34 | |
| CO ₂ | 34 | |

The data used in the generation rate calculations is presented in Table II. The ion pair generation rate, g , is normalized for 1 mA/cm^2 primary electron beam current at 100 keV and for $1 \text{ gas atom or molecule/cm}^3$.

It was calculated by dividing the value of the stopping power of the gas (taken from Berger¹⁶) by w , the average energy loss per ion pair. Also listed are the stopping powers¹⁶ and the ionisation cross sections for the production of ion pairs directly by the primary electrons. The generation rates in Table I were obtained by multiplying g by the beam current (in mA) and the fill gas number density.

3.2. Ionization by Electrons having about 1 eV Thermal Energy. The electrons in the plasma, produced by the ionizing processes described above, are maintained at an average energy of about 1 eV. The motion of these electrons is primarily random with a relatively slow drift ($\sim 10^6$ cm/s) toward the positive electrode. Because of the preponderance of electron-neutral collisions (compared to electron-electron collisions) and the non-thermal conditions (gas kinetic energy ~ 0.03 eV), the electron velocity distribution is non-Maxwellian (see References 8 and 9).

The correct method for the calculation of the ionization rate caused by these plasma electrons would be to use the velocity distribution appropriate to the particular gas mixture and the applied electric field. A much simpler procedure used here, which should yield rates within a factor of about two, is to take the Maxwellian velocity distribution equivalent in temperature to 2/3 of the average electron energy. The equation for $F(T_e, j)$ is

$$F(T_e, j) = \int_0^{\infty} \sigma(j, s) s f(s) ds \quad (14)$$

where j denotes the type of atom or molecule, s and $f(s)$ are the electron speed and speed distribution function and $\sigma(j, s)$ is the ionization cross section. The values for σ were taken from D. Rapp and P. Englander-Golden.¹⁹ The results of this integration are plotted in Figure 10 as a function

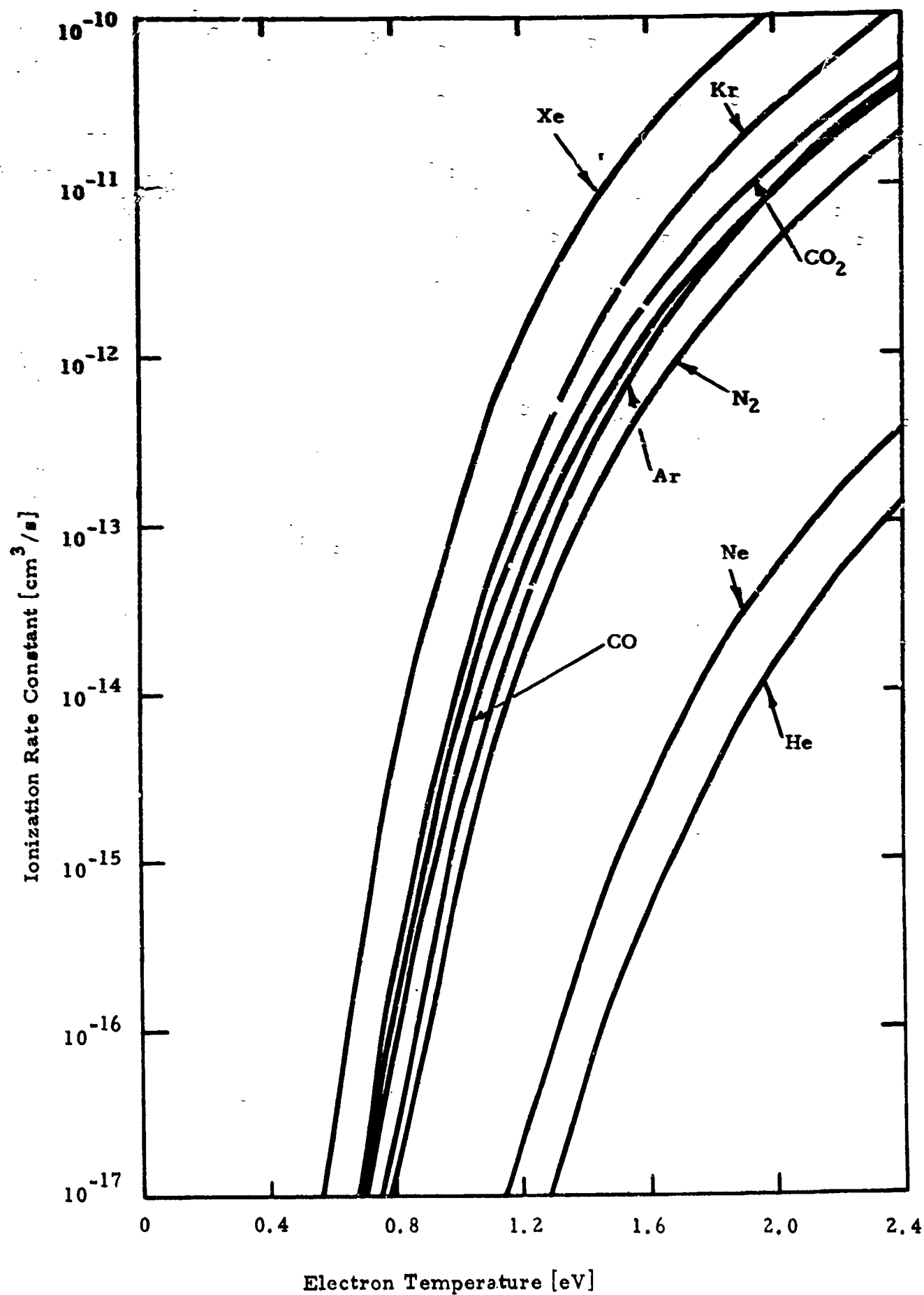
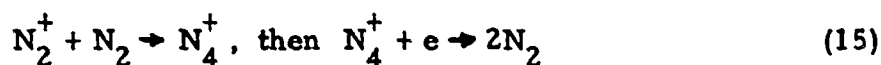


Figure 10. Ionization rate constant versus electron temperature.

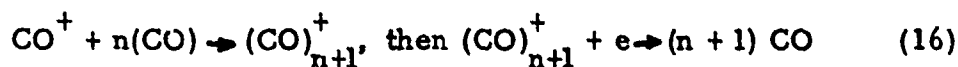
of electron temperature. In the above treatment only direct two body ionizing collisions are considered and ionization via metastable states or other processes are ignored.

3.3 Electron-Ion and Ion-Ion Recombination. The electron-ion recombination process represents the reverse of the electron impact ionization process. The ion-ion recombination process is responsible for the neutralization of the negative ions generated by the electron attachment process. The recombination processes are characterized by a rate coefficient α . The number of electron-ion recombination events per cm^3sec is given by $\alpha(j^+)n(j^+)n_e$, where $\alpha(j^+)$ depends on the ion species. For ion-ion recombination, α depends on both ion species. For example, for O_2^- and N_2^+ , the recombination rate is $\alpha(\text{O}_2^-, \text{N}_2^+)n(\text{O}_2^-)n(\text{N}_2^+)$. These two different recombination processes will be discussed separately.

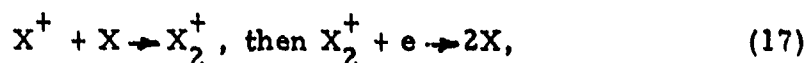
Of the many different types of electron-ion recombination processes possible (see, for example, McDaniel²⁰), it appears that, at gas pressures of the order of 1 atmosphere, dissociative recombination dominates. In particular, for diatomic molecules for example,



and



seem to be the important processes.^{10, 21} For the noble gases, a similar process accounts for the recombination,



where X represents one of the noble gas atoms. The dependence of some of the recombination processes on electron energy have been observed¹⁰ and the values quoted in Table III are for the temperatures indicated. The author is not aware of a value of α for electron-carbon dioxide recombination. In order to make the calculations, we have used the measured value of α for electron-carbon monoxide recombination. References to numerous other measurements of α are given by Oskam and Mittelstadt²⁴ and Hasted.²⁵

The author is not aware of ion-ion recombination coefficients for negative oxygen ions and positive carbon monoxide or carbon dioxide ions. Sayers²² has measured α for oxygen and nitrogen. Because of the similarity between CO and N₂ in the ionization and electron recombination processes we have assumed an equal recombination coefficient. For the lack of a better value we have also assumed the same α for CO₂.

3.4 Electron Attachment to and Detachment from Oxygen. The electrons in the discharge have a propensity to attach themselves to any oxygen molecules present in the system. In the process of attachment, the electron and oxygen molecule evolve energy (approximately 1.5 eV for O⁻ and 0.4 eV for O₂⁻) which has to be removed some way. We would then expect to have a variety of processes for attachment (as is the case for recombination). Phenomenologically these processes are divided into two classes: two body attachment and three body attachment. One may then write the rate of electron attachment as:

$$\beta' n_e = n_e n(\text{O}_2) [\beta + \sum_l K(l) n(l)] \quad (18)$$

where the l represents a particle species and the sum is to be extended over all the species present. β and K are the two and three body attachment coefficients respectively. These coefficients depend strongly on electron temperature as discussed in references 11 and 26. For a gas mixture of helium, nitrogen, carbon monoxide, and oxygen Eq. (18) becomes

TABLE III

Electron-Ion Recombination Coefficients

| Gas | α (cm ³ /s) | Remarks |
|--------------------------------|-------------------------------|--|
| He ₂ ⁺ | $4 \cdot 10^{-9}$ | At ~50 torr and T \approx .03 eV ²⁴ |
| Ne ₂ ⁺ | $3.4 \cdot 10^{-7}$ | At ~20 torr and T \approx .03 eV ²³ |
| Ar ₂ ⁺ | $8.8 \cdot 10^{-7}$ | At ~20 torr and T \approx .03 eV ²³ |
| Kr ₂ ⁺ | $1.2 \cdot 10^{-6}$ | At ~20 torr and T \approx .03 eV ²⁴ |
| Xe ₂ ⁺ | $1.4 \cdot 10^{-6}$ | At ~20 torr and T \approx .03 eV ²⁴ |
| N ₄ ⁺ | $3 \cdot 10^{-8}$ | At 1 atm and T \approx 1 eV ¹⁰ |
| (CO) _n ⁺ | $1 \cdot 10^{-7}$ | At 1 atm and T \approx 1 eV ¹⁰ |

Ion-Ion Recombination Coefficient

| | | |
|-----|------------------------|------------------------|
| Air | $\sim 1 \cdot 10^{-6}$ | At 1 atm ²² |
|-----|------------------------|------------------------|

$$\beta' = n_e n(O_2) [\beta + K(He)n(He) + K(N_2)n(N_2) + K(CO)n(CO) + K(O_2)n(O_2)]$$

The values of β' used in Figures 4, 6 and 7 were calculated from Eq. (13) and the data shown below:

$$\beta = 2 \times 10^{-13} \text{ cm}^3/\text{s}, \text{ extrapolated from Figure 8 of Reference 26 for } T_e = 1 \text{ eV}$$

$$K(O_2) = 1.5 \times 10^{-30} \text{ cm}^6/\text{s}, \text{ extrapolated from Figure 12 of Reference 26 for } T_e = 1 \text{ eV}$$

$$K(He) = 12^{-33} \text{ cm}^6/\text{s}, \text{ extrapolated from Figure 12 of Reference 26 for } T_e = 1 \text{ eV}$$

$$K(N_2) = 9 \times 10^{-33} \text{ cm}^6/\text{s}, \text{ extrapolated from Figure 12 of Reference 26 for } T_e = 1 \text{ eV.}$$

The "K" values for CO and CO₂ are unknown to the author and are therefore omitted here.

The reverse process, namely detachment by collision between O₂⁻ and O₂, appear to proceed at a very low rate at a temperature less than 400°K and at densities of O₂⁻ less than 10¹³/cm³, so that it may be neglected. The detachment rate at 400°K is 10⁻¹⁶ n(O₂) n(O₂⁻), according to Reference 26, page 411.

The attachment of electrons to CO appears to be unimportant at low electron temperatures. At the present, the author's only information is in Reference 11, which does not give data for electron energies below 9 eV. The attachment rate for CO₂, however, proceeds quite fast at electron energies of 4 eV.

3.5 Charge Transfer Processes. Exothermic charge transfer processes among the ions and molecules in a gas discharge proceed rapidly. The rate constants, k , for many of the expected processes have been measured and are given in Table IV. In order to help sort out the rates of these processes we indicate a characteristic lifetime for the ion which appears on the left side of the reaction equation. For example, the rate equation for the helium-nitrogen reaction is

$$\frac{dn(\text{He}^+)}{dt} = -kn(\text{He}^+)n(\text{N}_2).$$

We define the characteristic lifetime for the helium ion to be $(k n(\text{N}_2))^{-1}$. The characteristic times given in Table IV assume a total gas pressure of 760 torr at 273°K. and a mixture ratio of 3/2/1 for He, N₂ and CO (or CO₂) and 1 torr of O₂.

Cluster formation rates for the N₄⁺ molecule are also listed in Table IV, and for comparison, some recombination rates for electron-ion and ion-ion processes are included. For the electron-ion recombination characteristic times, an electron density of 10¹²/cm³ is assumed and, for the ion-ion process a nitrogen ion density of 10¹²/cm³ is assumed. The identity of the recombining oxygen and nitrogen ions is not known and therefore the final equation of Table IV represents the process schematically only.

The characteristic lifetimes listed in Table IV show that the main ionic charge transfer processes proceed very fast compared to the ion-electron recombination processes.

TABLE IV. Rates for some Molecular Ion Processes

| Reaction | k (cm ³ /s) | Reference Number | Characteristic Lifetime |
|--|---|------------------|-------------------------|
| <u>Charge Exchange</u> | | | |
| He ⁺ + N ₂ → He + N + N ⁺ | 1.7 × 10 ⁻⁹ | 27 | 0.6 × 10 ⁻¹⁰ |
| He ⁺ + CO → He + O + C ⁺ | 1.7 × 10 ⁻⁹ | 27 | 1.3 × 10 ⁻¹⁰ |
| He ⁺ + CO ₂ → { He + CO + O ⁺ He + CO ⁺ + O } | 1.2 × 10 ⁻⁹ | 27 | 1.8 × 10 ⁻¹⁰ |
| N ₂ ⁺ + { CO } → { N ₂ + CO ⁺ N ₂ + CO ₂ ⁺ } | 0.7 × 10 ⁻⁹ | 28 | 3.1 × 10 ⁻¹⁰ |
| | 0.9 × 10 ⁻⁹ | 28 | 2.4 × 10 ⁻¹⁰ |
| N ⁺ + { CO } → { N + CO ⁺ N + CO ₂ ⁺ } | 0.9 × 10 ⁻⁹ | 28 | 2.4 × 10 ⁻¹⁰ |
| | 1.3 × 10 ⁻⁹ | 28 | 1.7 × 10 ⁻¹⁰ |
| <u>Cluster Formation</u> | | | |
| N ₂ ⁺ + N ₂ → N ₄ ⁺ | 6 × 10 ⁻¹¹ | 29 | 18 × 10 ⁻¹⁰ |
| CO ⁺ + n(CO) → (CO) _{n+1} ⁺ | ? Probably similar to N ₂ ⁺ + N ₂ reaction | | |
| <u>Recombination</u> | | | |
| He ₂ ⁺ + e → 2He | ~ 10 ⁻⁹ | | |
| (CO) _n ⁺ + e → nCO | ~ 10 ⁻⁷ | | |
| N ₄ ⁺ + e → 2N ₂ | ~ 10 ⁻⁷ | | |
| O _x ⁺ + N _y ⁺ → O _x + N _y | ~ 10 ⁻⁶ | | |

4.0 SUMMARY

In an electrical discharge gas laser, the two most important plasma parameters for electron pumping are electron density and electron energy. In this report the important physical processes which control the electron density are discussed and a set of equations for calculating the density are formulated and solved. It is shown that the equilibrium value of the electron density of an electron beam stabilized CO or CO₂ laser discharge may be calculated approximately from a simple quadratic equation. Because of rapid charge transfer, the only significant number of positive ions present in the discharge are those of CO or CO₂. The electron generation rate is the sum of the rates for all the various gas constituents. These generation rates are proportional to the electron beam current density, and mass density of the fill gas. The calculated electron densities from these formulations are in reasonable agreement with measured values.

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